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# PATENT ABSTRACTS OF JAPAN

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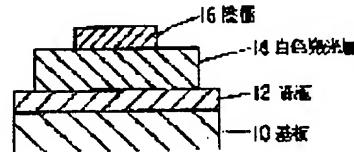
(21)Application number : 08-047991	(71)Applicant : CHISSO CORP HOKURIKU ELECTRIC IND CO LTD
(22)Date of filing : 09.02.1996	(72)Inventor : FURUKAWA KENJI IZUMISAWA YUSHO FUKUMOTO SHIGERU TANPO TETSUYA

## (54) ORGANIC THIN FILM ELECTROLUMINESCENT ELEMENT AND ITS MANUFACTURE

### (57)Abstract:

**PROBLEM TO BE SOLVED:** To provide an organic thin film electroluminescent element with long time stability and simplify the process of evaporation by mixing several organic electroluminescent materials together to form a white emissive one on an anode with flash evaporation.

**SOLUTION:** An organic thin film electroluminescent element has an anode 12 as an ITO transparent conductive layer formed on the surface of a glass, transparent resin or silica substrate 10 and a Mg cathode 16 formed outside thereof. A white emissive layer 14 is preferable made of base material with a band gap of 2.6 or more to combine charge transportation material and emission material. The emission material is formed by mixing several types of organic electroluminescent materials emitting red, green, blue or at least red or blue based fluorescent light. For flash evaporation, the organic electroluminescent materials mixed together at a preset ratio are dropped on an evaporating source heated to about 300–600°C and evaporated without rest.



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## CLAIMS

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[Claim(s)]

[Claim 1] Organic thin film electroluminescence devices by which the anode plate was formed on the substrate, two or more kinds of organic electroluminescence ingredients of the organic electroluminescence ingredient of a blue system and the organic electroluminescence ingredient of a red system were mixed at least by the front face, and the luminous layer of the monolayer for which light can be emitted white was formed in it with flash plate vacuum deposition.

[Claim 2] The above-mentioned organic electroluminescence ingredients are organic thin film electroluminescence devices according to claim 1 which are what serves both as a charge transport ingredient and luminescent material.

[Claim 3] Organic thin film electroluminescence devices according to claim 1 with which an organic electroluminescence ingredient and at least one kind of charge transport ingredient are mixed by the above-mentioned white luminous layer.

[Claim 4] The manufacture approach of the organic thin film electroluminescence devices which vapor-deposit the above-mentioned mixed luminescent material on the front face with flash plate vacuum deposition, form a white luminous layer in it by mixing two or more kinds of organic electroluminescence ingredients of the organic electroluminescence ingredient of a blue system, and the organic electroluminescence ingredient of a red system at least, forming a mixed luminescent material for which light can be emitted white, and forming an anode plate on a substrate, and establish cathode in the front face.

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## DETAILED DESCRIPTION

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### [Detailed Description of the Invention]

#### [0001]

[Field of the Invention] This invention is the electroluminescence devices by the organic compound of the thin film prepared on the substrate, and relates to the organic thin film electroluminescence devices used for the flat-surface light source or a display, and its manufacture approach.

#### [0002]

[Description of the Prior Art] When it was going to obtain white luminescence conventionally in organic thin film electroluminescence devices (an organic EL device is called below), as shown in drawing 6, each luminous layers 3, 4, and 5 of red, green, and blue were formed in the front face of the anode plate 2 formed in the front face of a substrate 1 in order, cathode 6 was formed in the outside, and white luminescence had been obtained. Each of these luminous layers 3, 4, and 5 form membranes with vacuum deposition respectively.

[0003] Moreover, the organic material which forms each luminescent color is made to vapor-deposit simultaneously from the separate source of vacuum evaporationo respectively, and there are also what shortened the vacuum evaporationo process, and the approach of forming a white luminous layer in an anode plate front face by melting the above-mentioned organic material to an organic solvent, and being applied or immersed.

[0004] Furthermore, as shown in drawing 7, the electron hole transporting bed 7 is formed on the surface of an anode plate, and the white luminescence EL element which carries out vacuum evaporationo formation of the luminous layer 8 of single ingredients, such as a benzothiazole Zn derivative, on it is also proposed.

#### [0005]

[Problem(s) to be Solved by the Invention] What vapor-deposits each luminous layers 3, 4, and 5 of the above-mentioned Prior art in order, and forms membranes had many counts of vacuum evaporationo, its process was complicated, and, moreover, it was that from which an organic electroluminescence ingredient is weak with heat, and the property of the EL element done with the heat at the time of vacuum evaporationo does not vary, or the desired engine performance is not obtained. Moreover, when evaporating the organic electroluminescence ingredient of each color from the source of vacuum evaporationo independently simultaneously, it was difficult for control of the presentation ratio of each organic electroluminescence ingredient at the time of membrane formation to form membranes into a difficult ratio predetermined to accuracy.

[0006] Moreover, when mixing the organic electroluminescence ingredient of each color, melting to an organic solvent and carrying out spreading etc., it was easy to mix an impurity with a solvent, and what has good quality was what is not obtained. Furthermore, although shown in drawing 7, the count of vacuum evaporationo is required also for a case twice [ at least ], a process becomes complicated too, and the adverse effect to each organic electroluminescence ingredient by heat is also produced.

[0007] This invention is an easy production process and aims at offering the organic thin film electroluminescence devices which can obtain white luminescence of high quality to stability, and its

manufacture approach.

[0008]

[Means for Solving the Problem] This invention is the organic thin film electroluminescence devices by which the anode plate was formed, two or more kinds of organic electroluminescence ingredients which take out the fluorescence of the long wavelength of a red system from the short wavelength of a blue system to that front face respectively were mixed, and the luminous layer of the monolayer for which light can be emitted white was formed with flash plate vacuum deposition on the substrate. Moreover, at least one side of the above-mentioned organic electroluminescence ingredient serves both as a charge transport ingredient and luminescent material. Or they are the organic thin film electroluminescence devices with which each above-mentioned organic electroluminescence ingredient and at least one kind of charge transport ingredient are mixed by the above-mentioned white luminous layer.

[0009] Moreover, this invention is the manufacture approach of the organic thin film electroluminescence devices which vapor-deposit the above-mentioned mixed luminescent material on that front face with flash plate vacuum deposition, form a white luminous layer in it by mixing two or more kinds of organic electroluminescence ingredients of the organic electroluminescence ingredient of a blue system, and the organic electroluminescence ingredient of a red system at least, forming a mixed luminescent material for which light can be emitted white, and forming an anode plate on a substrate, and establish cathode in that front face.

[0010]

[Embodiment of the Invention] Hereafter, 1 operation gestalt of this invention is explained based on a drawing. As the organic thin film electroluminescence devices of this operation gestalt are shown in drawing 1, the anode plate 12 which are transparent conductive layers, such as ITO, is formed in the front face of the substrates 10, such as glass, transparency resin, and a quartz, the white luminous layer 14 is formed in that front face, and cathode 16 is formed in that outside with Mg etc.

[0011] The thing 2.6eV or more of a band gap is [ the white luminous layer 14 / a parent ingredient ] good. As an electronic transport ingredient, they are a distyrylbiphenyl derivative, an OKISA diazole derivative, a bis-CHIRIRU anthracene derivative, a benzo oxazole thiophene derivative, etc. among this parent ingredient. As a hole transport ingredient, there are a triphenylamine derivative, a hydrazone derivative, an arylamine derivative, etc. Moreover, the ratio of the above-mentioned electronic transport ingredient and a hole transport ingredient can be suitably changed in 10:90 thru/or 90:10.

[0012] As a luminescent material added into a parent ingredient, a coumarin derivative, the Quinacridone derivative, a fluoro SEIN derivative, etc. can use the organic pigment which gives off the fluorescence of a dicyanomethylene derivative, a phenoxyazone derivative, a squarylium derivative, etc. as a red luminescent material as a blue luminescent material as green luminescent material, such as a tetra-phenyl butadiene derivative, an anthracene derivative, and a perylene derivative. Moreover, a distyrylbiphenyl derivative serves also as blue luminescent material. The presentation ratio of the above-mentioned parent ingredient and the above-mentioned add-in material can be suitably changed in 100:0.01 thru/or 100:20.

[0013] The manufacture approach of the organic thin film electroluminescence devices of this operation gestalt mixes each color of red, green, and blue, or two or more kinds of organic electroluminescence ingredients which emit the fluorescence of reddish and a blue system at least, and forms a mixed luminescent material. And an anode plate 16 is formed by vacuum evaporation etc. on a substrate 10, the above-mentioned mixed luminescent material is quickly vapor-deposited on the front face with flash plate vacuum deposition, and the white luminous layer 14 is formed in it. And cathode 12 is established in the front face by vacuum evaporation etc.

[0014] Flash plate vacuum deposition drops 300-600 degrees C of organic electroluminescence ingredients beforehand mixed by the predetermined ratio in the source of vacuum evaporation preferably heated at 400-500 degrees C, and evaporates an organic electroluminescence ingredient at a stretch. Moreover, the organic electroluminescence ingredient is held into a container, the container is heated quickly, and it may be made to vapor-deposit at a stretch.

[0015]

## [Example]

an example 1 -- in this example, ITO was formed as an anode plate 12, the distyrylbiphenyl derivative which carries out blue luminescence as a triphenylamine derivative and an electronic transport ingredient as an electron hole transport ingredient of the white luminous layer 14 was mixed at a rate of 1:2 by the mole ratio, and the parent ingredient was formed. And the dicyanomethylene derivative was blended at a weight rate of 0.1 to the above-mentioned parent ingredient 100 as a red luminescent material. This mixed luminescent material was formed in the thickness of 1000A as a white luminous layer 14. Next, Mg was used for cathode 16.

[0016] A degree of vacuum is  $6 \times 10^{-6}$  Torr, and formed vacuum evaporationo conditions with the evaporation rate of 50A/sec. The source of vacuum evaporationo was raised to 400 degrees C within 20 seconds.

[0017] The organic EL device obtained according to this example shows a current density-luminescence brightness property as shown an applied-voltage-luminescence brightness property as shown in drawing 2 and shown in drawing 3 . Furthermore, emission spectrum distribution as shown in drawing 4 is shown. Moreover, although the reinforcement of the wavelength near 550nm is high and the white which green cut slightly is shown by this spectrum, better white luminescence can also be obtained by choosing red and a blue fluorescence ingredient suitably.

[0018] an example 2 -- in this example, phenoxazone induction was blended with the same above-mentioned parent ingredient as the above-mentioned example 1 at a weight rate of 0.1 to the above-mentioned parent ingredient 100 as a red luminescent material. In addition, membrane formation conditions are the same as the above-mentioned example 1.

[0019] Thereby, the white light of the emission spectrum distribution shown as the continuous line of drawing 5 was acquired. Moreover, the luminescence brightness in this case was weak as compared with a maximum of 250 cd/m<sup>2</sup> and the above-mentioned example 1.

[0020] an example 3 -- phenoxazone induction and a coumarin derivative were blended with the same above-mentioned parent ingredient as the above-mentioned example 1 at a weight rate of 0.1 and 0.3 to the abcve-mentioned parent ingredient 100 also by this example as a red luminescent material. In addition, membrane formation conditions are the same as the above-mentioned example 1.

[0021] The white light of the emission spectrum distribution shown with the broken line of drawing 5 was acquired by this, the luminescence reinforcement near [ which had fallen in the example 2 ] 530nm could be raised, and the white light which spreads widely in a visible region was able to be acquired. Moreover, the luminescence brightness in this case was also weak as compared with a maximum of 250 cd/m<sup>2</sup> and the above-mentioned example 1.

[0022]

[Effect of the Invention] Since the organic EL device of this invention forms on an anode plate what was mixed and enabled white luminescence of an organic electroluminescence ingredient by flash plate vacuum evaporationo, vacuum evaporationo is performed for a short time, it is very rare for an organic electro uminescence ingredient to receive an adverse effect with heat, and a good property is acquired. [ two or more ] And a vacuum evaporationo process is simplified and a white luminescence EL element can be formed easily.

[0023] Moreover, vacuum evaporationo equipment can also be miniaturized and complicated adjustment of control of a presentation, the membrane formation rate at the time of membrane formation, etc. is unnecessary. Furthermore, although an organic layer is a monolayer, and component degradation arises by the interaction which is an organic layer when an organic layer is that by which the laminating was carried out, with the component of this invention, this does not arise and a component operates to stability for a long period of time.

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#### TECHNICAL FIELD

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[Field of the Invention] This invention is the electroluminescence devices by the organic compound of the thin film prepared on the substrate, and relates to the organic thin film electroluminescence devices used for the flat-surface light source or a display, and its manufacture approach.

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PRIOR ART

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[Description of the Prior Art] When it was going to obtain white luminescence conventionally in organic thin film electroluminescence devices (an organic EL device is called below), as shown in drawing 6 , each luminous layers 3, 4, and 5 of red, green, and blue were formed in the front face of the anode plate 2 formed in the front face of a substrate 1 in order, cathode 6 was formed in the outside, and white luminescence had been obtained. Each of these luminous layers 3, 4, and 5 form membranes with vacuum deposition respectively.

[0003] Moreover, the organic material which forms each luminescent color is made to vapor-deposit simultaneously from the separate source of vacuum evaporation respectively, and there are also what shortened the vacuum evaporation process, and the approach of forming a white luminous layer in an anode plate front face by melting the above-mentioned organic material to an organic solvent, and being applied or immersed.

[0004] Furthermore, as shown in drawing 7 , the electron hole transporting bed 7 is formed on the surface of an anode plate, and the white luminescence EL element which carries out vacuum evaporation formation of the luminous layer 8 of single ingredients, such as a benzothiazole Zn derivative, on it is also proposed.

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EFFECT OF THE INVENTION

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[Effect of the Invention] Since the organic EL device of this invention forms on an anode plate what was mixed and enabled white luminescence of an organic electroluminescence ingredient by flash plate vacuum evaporationo, vacuum evaporationo is performed for a short time, it is very rare for an organic electroluminescence ingredient to receive an adverse effect with heat, and a good property is acquired. [ two or more ] And a vacuum evaporationo process is simplified and a white luminescence EL element can be formed easily.

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TECHNICAL PROBLEM

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[0007] This invention is an easy production process and aims at offering the organic thin film electroluminescence devices which can obtain white luminescence of high quality to stability, and its manufacture approach.

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## MEANS

[Means for Solving the Problem] This invention is the organic thin film electroluminescence devices by which the anode plate was formed, two or more kinds of organic electroluminescence ingredients which take out the fluorescence of the long wavelength of a red system from the short wavelength of a blue system to that front face respectively were mixed, and the luminous layer of the monolayer for which light can be emitted white was formed with flash plate vacuum deposition on the substrate. Moreover, at least one side of the above-mentioned organic electroluminescence ingredient serves both as a charge transport ingredient and luminescent material. Or they are the organic thin film electroluminescence devices with which each above-mentioned organic electroluminescence ingredient and at least one kind of charge transport ingredient are mixed by the above-mentioned white luminous layer.

[0009] Moreover, this invention is the manufacture approach of the organic thin film electroluminescence devices which vapor-deposit the above-mentioned mixed luminescent material on that front face with flash plate vacuum deposition, form a white luminous layer in it by mixing two or more kinds of organic electroluminescence ingredients of the organic electroluminescence ingredient of a blue system, and the organic electroluminescence ingredient of a red system at least, forming a mixed luminescent material for which light can be emitted white, and forming an anode plate on a substrate, and establish cathode in that front face.

[0010]

[Embodiment of the Invention] Hereafter, 1 operation gestalt of this invention is explained based on a drawing. As the organic thin film electroluminescence devices of this operation gestalt are shown in drawing 1, the anode plate 12 which are transparent conductive layers, such as ITO, is formed in the front face of the substrates 10, such as glass, transparency resin, and a quartz, the white luminous layer 14 is formed in that front face, and cathode 16 is formed in that outside with Mg etc.

[0011] The thing 2.6eV or more of a band gap is [ the white luminous layer 14 / a parent ingredient ] good. As an electronic transport ingredient, they are a distyrylbiphenyl derivative, an OKISA diazole derivative, a bis-CHIRIRU anthracene derivative, a benzo oxazole thiophene derivative, etc. among this parent ingredient. As a hole transport ingredient, there are a triphenylamine derivative, a hydrazone derivative, an arylamine derivative, etc. Moreover, the ratio of the above-mentioned electronic transport ingredient and a hole transport ingredient can be suitably changed in 10:90 thru/or 90:10.

[0012] As a luminescent material added into a parent ingredient, a coumarin derivative, the Quinacridone derivative, a fluoro SEIN derivative, etc. can use the organic pigment which gives off the fluorescence of a dicyanomethylene derivative, a phenoxyzone derivative, a squarylium derivative, etc. as a red luminescent material as a blue luminescent material as green luminescent material, such as a tetra-phenyl butadiene derivative, an anthracene derivative, and a perylene derivative. Moreover, a distyrylbiphenyl derivative serves also as blue luminescent material. The presentation ratio of the above-mentioned parent ingredient and the above-mentioned add-in material can be suitably changed in 100:0.01 thru/or 100:20.

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ingredients which emit the fluorescence of reddish and a blue system at least, and forms a mixed luminescent material. And an anode plate 16 is formed by vacuum evaporation etc. on a substrate 10, the above-mentioned mixed luminescent material is quickly vapor-deposited on the front face with flash plate vacuum deposition, and the white luminous layer 14 is formed in it. And cathode 12 is established in the front face by vacuum evaporation etc.

[0014] Flash plate vacuum deposition drops 300-600 degrees C of organic electroluminescence ingredients beforehand mixed by the predetermined ratio in the source of vacuum evaporation preferably heated at 400-500 degrees C, and evaporates an organic electroluminescence ingredient at a stretch. Moreover, the organic electroluminescence ingredient is held into a container, the container is heated quickly, and it may be made to vapor-deposit at a stretch.

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## EXAMPLE

## [Example]

an example 1 -- in this example, ITO was formed as an anode plate 12, the distyrylbiphenyl derivative which carries out blue luminescence as a triphenylamine derivative and an electronic transport ingredient as an electron hole transport ingredient of the white luminous layer 14 was mixed at a rate of 1:2 by the mole ratio, and the parent ingredient was formed. And the dicyanomethylene derivative was blended at a weight rate of 0.1 to the above-mentioned parent ingredient 100 as a red luminescent material. This mixed luminescent material was formed in the thickness of 1000A as a white luminous layer 14. Next, Mg was used for cathode 16.

[0016] A degree of vacuum is  $6 \times 10^{-6}$  Torr, and formed vacuum evaporation conditions with the evaporation rate of 50A/sec. The source of vacuum evaporation was raised to 400 degrees C within 20 seconds.

[0017] The organic EL device obtained according to this example shows a current density-luminescence brightness property as shown an applied-voltage-luminescence brightness property as shown in drawing 2 and shown in drawing 3 . Furthermore, emission spectrum distribution as shown in drawing 4 is shown. Moreover, although the reinforcement of the wavelength near 550nm is high and the white which green cut slightly is shown by this spectrum, better white luminescence can also be obtained by choosing red and a blue fluorescence ingredient suitably.

[0018] an example 2 -- in this example, phenoxazone induction was blended with the same above-mentioned parent ingredient as the above-mentioned example 1 at a weight rate of 0.1 to the above-mentioned parent ingredient 100 as a red luminescent material. In addition, membrane formation conditions are the same as the above-mentioned example 1.

[0019] Thereby, the white light of the emission spectrum distribution shown as the continuous line of drawing 5 was acquired. Moreover, the luminescence brightness in this case was weak as compared with a maximum of 250 cd/m<sup>2</sup> and the above-mentioned example 1.

[0020] an example 3 -- phenoxazone induction and a coumarin derivative were blended with the same above-mentioned parent ingredient as the above-mentioned example 1 at a weight rate of 0.1 and 0.3 to the above-mentioned parent ingredient 100 also by this example as a red luminescent material. In addition, membrane formation conditions are the same as the above-mentioned example 1.

[0021] The white light of the emission spectrum distribution shown with the broken line of drawing 5 was acquired by this, the luminescence reinforcement near [ which had fallen in the example 2 ] 530nm could be raised, and the white light which spreads widely in a visible region was able to be acquired. Moreover, the luminescence brightness in this case was also weak as compared with a maximum of 250 cd/m<sup>2</sup> and the above-mentioned example 1.

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#### DESCRIPTION OF DRAWINGS

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[Brief Description of the Drawings]

[Drawing 1] It is the sectional view of the organic thin film electroluminescence devices of 1 operation gestalt of this invention.

[Drawing 2] It is the graph which shows the applied-voltage-luminescence brightness property of the example 1 of the organic thin film electroluminescence devices by this invention.

[Drawing 3] It is the graph which shows the current density-luminescence brightness property of the example 1 of the organic thin film electroluminescence devices by this invention.

[Drawing 4] It is the graph which shows emission spectrum distribution of the example 1 of the organic thin film electroluminescence devices by this invention.

[Drawing 5] It is the graph which shows emission spectrum distribution of the example 2 of the organic thin film electroluminescence devices by this invention, and an example 3.

[Drawing 6] It is the sectional view of the organic thin film electroluminescence devices of a Prior art.

[Drawing 7] It is the sectional view of the organic thin film electroluminescence devices of other Prior arts.

[Description of Notations]

1 Ten Substrate

2 12 Anode plate

6 16 Cathode

14 White Luminous Layer

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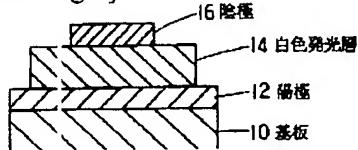
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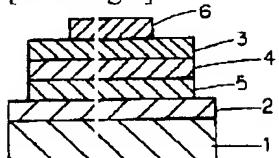
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## DRAWINGS

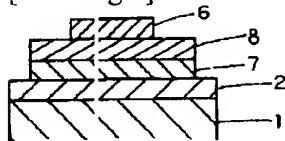
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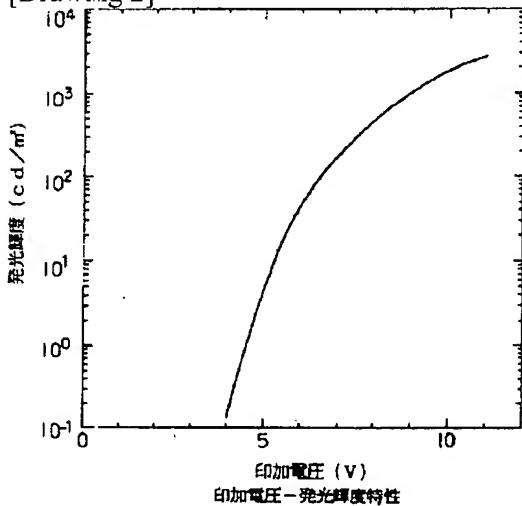
[Drawing 6]



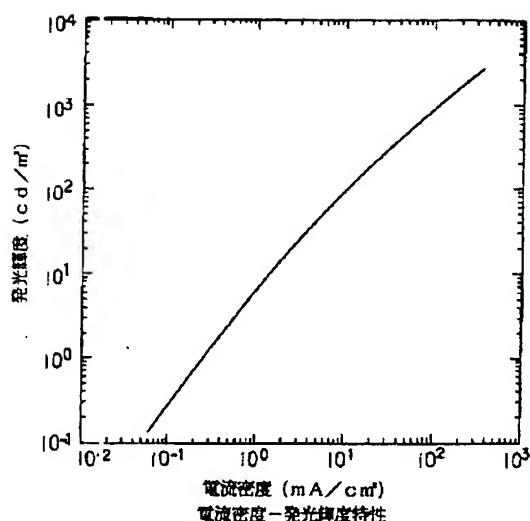
[Drawing 7]



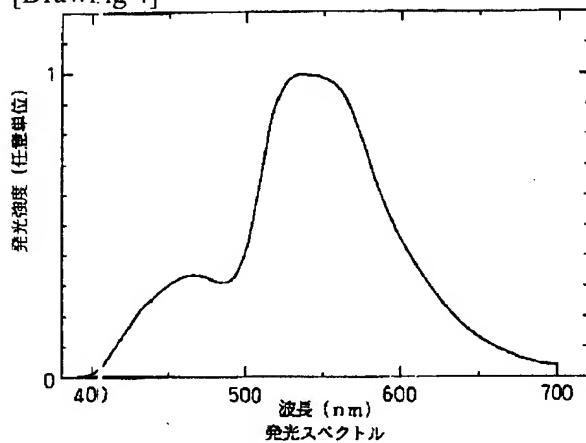
[Drawing 2]



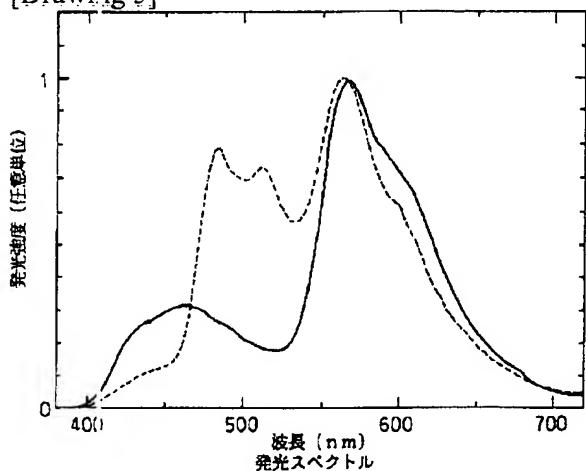
[Drawing 3]



[Drawing 4]



[Drawing 5]



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